

# Orbital Dzyaloshinskii-Moriya Exchange Interaction

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Superexchange calculation is performed for multi-orbital band models with broken inversion symmetry. Orbital-changing hopping terms allowed by the symmetry breaking electric field lead to a new kind of orbital exchange term closely resembling the Dzyaloshinskii-Moriya spin exchange. The final superexchange Hamiltonian in two dimensions is expressed in terms of Gell-Mann matrices that break down as three spin operators and five nematic (quadrupole) order operators. Mean-field phase diagram exhibits a rich structure including anti-ferro-orbital, ferro-orbital, and both single and multiple spiral-orbital phases. Superexchange calculation for spinful, two-orbital model also strongly suggests a robust chiral-orbital order in the ground state.

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Strong on-site repulsion transforms the Bloch bands into an insulator where the residual low-energy dynamics are the spin degrees of freedom interacting with each other via the superexchange mechanism [1]. For spin-orbit-coupled bands, the spin-flip hopping processes result in a new type of spin exchange called the Dzyaloshinskii-Moriya (DM) interaction [2] under the superexchange process. Symmetry-wise, local inversion symmetry breaking such as the bond distortion for a pair of adjacent magnetic orbitals, in addition to the spin-orbit interaction (SOI), is the pre-requisite for the DM interaction to make its appearance in a given system. The ordered magnetic ground state is modified from the simple collinear structure as a result of the DM exchange.

Inversion symmetry breaking (ISB) on the global scale takes place for surfaces and interfaces and affects the band structure directly with new effects such as the Rashba interaction [3]. The situation was recently reviewed carefully in Refs. [4, 5] where it was shown that the symmetry-breaking electric field along the  $z$ -direction modifies the band structure within the  $xy$ -plane by allowing previously forbidden hopping processes. Examples are  $p_{x(y)} \leftrightarrow p_z$  orbital hopping in the  $p$ -band, and  $d_{xy} \leftrightarrow d_{zx}$  orbital hopping in the  $t_{2g}$ -band. It was further shown [5] that the new hopping terms arising from ISB can be cast in the form  $-\gamma \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^\dagger \mathbf{L} \cdot (\mathbf{k} \times \hat{z}) \Psi_{\mathbf{k}}$  around the  $\Gamma$  ( $\mathbf{k} = 0$ ) point, where  $\Psi_{\mathbf{k}}$  is the collection of, say,  $p$ -orbital operators  $(X_{\mathbf{k}}, Y_{\mathbf{k}}, Z_{\mathbf{k}})^T$  in momentum coordinates  $\mathbf{k}$ ,  $\gamma$  is a parameter measuring the degree of ISB, and  $\mathbf{L}$  is the spin-1 orbital angular momentum (OAM) operator. Just from the form of the new interaction it is clear that each band will carry polarized OAM proportional to  $\hbar(\mathbf{k} \times \hat{z})$  with the respective helicities  $h = +1, 0, -1$  [5]. The enlargement of effective spin size from  $1/2$  (as in electrons' spin) to  $1$  (as in degenerate  $p$ -orbital bands) results in the appearance of the third band that remains unpolarized. The chiral structure of the OAM, dubbed the “orbital Rashba effect”, occurs even

in the complete absence of SOI and has been confirmed by circular dichroism ARPES in the weak-SOI material, Cu [6].

The new hopping processes allowed by ISB are the orbital analogues of spin-flip hoppings in spin-orbit-coupled bands. Therefore, the two necessary conditions for the emergence of spin-DM interaction - ISB and SOI - are automatically fulfilled when a symmetry-breaking electric field acts perpendicular to the two-dimensional surface. The purpose of this paper is to review this situation carefully in the limit of strong on-site interaction regime to ask if an orbital analogue of spin-DM interaction exists.

We address this question in two complementary ways. First, the spin-orbit effect will be ignored in favor of a spinless multi-orbital band model with strong on-site repulsion. The proposed model may find useful applications in spinless fermion lattice or in the bosonic cold-atom optical lattice problem as several recent papers on this subject can testify [7, 8]. The orbital version of DM interaction indeed arises from superexchange-type calculation, but with complications from the three-orbital rather than the two-spin degrees of freedom present at each site. The effective spin-1 model involves not only the usual spin-spin exchange among the spin-1 operators, but also the interaction among the nematic operators. As noted recently in the cold-atom context [8], the whole low-energy Hamiltonian is expressed in terms of the eight Gell-Mann matrices, which we further demonstrate to be decomposed rather nicely into a set of three spin matrices and the other five representing the quadrupole (nematic) order.

Assuming three  $p$ -orbital states (of either hard-core bosons or spinless fermions) at each site, we write down a square lattice Hamiltonian with nearest-neighbor hopping  $H_t = \sum_i (H_{i,i+\hat{x}} + H_{i,i+\hat{y}})$ ,

$$\begin{aligned}
H_{i,i+\hat{x}} &= t_a X_i^\dagger X_{i+\hat{x}} + t_b [Y_i^\dagger Y_{i+\hat{x}} + Z_i^\dagger Z_{i+\hat{x}}] \\
&\quad + \gamma (X_i^\dagger Z_{i+\hat{x}} - Z_i^\dagger X_{i+\hat{x}}) + h.c. \\
H_{i,i+\hat{y}} &= t_a Y_i^\dagger Y_{i+\hat{y}} + t_b [X_i^\dagger X_{i+\hat{y}} + Z_i^\dagger Z_{i+\hat{y}}] \\
&\quad + \gamma (Y_i^\dagger Z_{i+\hat{y}} - Z_i^\dagger Y_{i+\hat{y}}) + h.c. \quad (1)
\end{aligned}$$

Two hopping integrals  $t_a$  and  $t_b$  are given for  $\sigma$ - and  $\pi$ -bonding orbitals, respectively. Inter-orbital hopping becomes possible when the ISB parameter  $\gamma$  is nonzero [4, 5]. All hopping parameters are real due to the assumed time-reversal invariance. Three-component spinor can be formed,  $\psi_i = (X_i \ Y_i \ Z_i)^T$ , representing the  $p_x$ -,  $p_y$ -, and  $p_z$ -orbitals at the site  $i$ . To reduce the vast com-

plexity of the superexchange calculation to a manageable minimum we adopt a simplified multi-orbital Hubbard interaction  $H_U = U \sum_i n_i(n_i - 1)$ , where  $n_i = X_i^\dagger X_i + Y_i^\dagger Y_i + Z_i^\dagger Z_i$  is the total particle number at the site  $i$ .

Superexchange calculation at 1/3-filling (one particle per each site) can proceed with the Hamiltonian  $H = H_t + H_U$ . Without much loss of generality and to make the final outcome of the calculation tractable to follow (even to write down!), we had to further drop the weaker  $\pi$ -hopping term  $t_b$ . The exchange Hamiltonian thus obtained, writing  $U \equiv 1$  and  $t_a \equiv 2t$ , reads  $\mathcal{H} = \mathcal{H}_{\hat{x}} + \mathcal{H}_{\hat{y}}$ :

$$\begin{aligned}
\mathcal{H}_{\hat{x}} &= 2t^2 \sum_i (\lambda_i^x + [\lambda_i^x]^2 - 1)(\lambda_{i+\hat{x}}^x + [\lambda_{i+\hat{x}}^x]^2 - 1) + 2t\gamma \sum_i \left[ (\lambda_i^x + [\lambda_i^x]^2) \lambda_{i+\hat{x}}^4 - \lambda_i^4 (\lambda_{i+\hat{x}}^x + [\lambda_{i+\hat{x}}^x]^2) \right] \\
&\quad + \gamma^2 \sum_i \left[ \lambda_i^5 \lambda_{i+\hat{x}}^5 - \lambda_i^4 \lambda_{i+\hat{x}}^4 - \lambda_i^x \lambda_{i+\hat{x}}^x + ([\lambda_i^x]^2 - 1)([\lambda_{i+\hat{x}}^x]^2 - 1) \right], \\
\mathcal{H}_{\hat{y}} &= 2t^2 \sum_i (\lambda_i^y + [\lambda_i^y]^2 - 1)(\lambda_{i+\hat{y}}^y + [\lambda_{i+\hat{y}}^y]^2 - 1) + 2t\gamma \sum_i \left[ (\lambda_i^y + [\lambda_i^y]^2) \lambda_{i+\hat{y}}^6 - \lambda_i^6 (\lambda_{i+\hat{y}}^y + [\lambda_{i+\hat{y}}^y]^2) \right] \\
&\quad + \gamma^2 \sum_i \left[ \lambda_i^7 \lambda_{i+\hat{y}}^7 - \lambda_i^6 \lambda_{i+\hat{y}}^6 - \lambda_i^y \lambda_{i+\hat{y}}^y + ([\lambda_i^y]^2 - 1)([\lambda_{i+\hat{y}}^y]^2 - 1) \right]. \quad (2)
\end{aligned}$$

Here  $\lambda^\alpha$  are the Gell-Mann matrices,  $\alpha = 1 \cdots 8$ , and  $\lambda^x = \text{diag}(1, 0, -1)$ ,  $\lambda^y = \text{diag}(0, 1, -1)$  are combinations of  $\lambda^3$ ,  $\lambda^8$  and the unit matrix.

Although the Hamiltonian obtained above appears very complex,  $\mathcal{H}_{\hat{x}}$  or  $\mathcal{H}_{\hat{y}}$  examined in isolation has a simple interpretation. Note that  $\mathcal{H}_{\hat{x}}$  only involves the three operators ( $\lambda^4, \lambda^5, \lambda^x$ ) that, in common, have zero matrix elements for the  $p_y$ -orbital state and reduce to the three Pauli matrices  $\boldsymbol{\tau} = (\tau^x, \tau^y, \tau^z)$  within the  $(p_x, p_z)$ -orbital subspace. In particular the part of  $\mathcal{H}_{\hat{x}}$  involving the first power of  $\gamma$  can be re-written as

$$2t\gamma \sum_i [\tau_i^z \tau_{i+\hat{x}}^x - \tau_i^x \tau_{i+\hat{x}}^z] = 2t\gamma \sum_i \hat{y} \cdot [\boldsymbol{\tau}_i \times \boldsymbol{\tau}_{i+\hat{x}}], \quad (3)$$

which is exactly the orbital analogue of the DM spin exchange, or “orbital DM” (ODM) exchange. The real-valued transition amplitudes obtained from the superexchange process excludes the imaginary  $\tau_y$  operator, permitting  $\hat{y} \cdot [\boldsymbol{\tau}_i \times \boldsymbol{\tau}_{i+\hat{x}}]$  as the only permissible DM interaction. The  $\gamma^2$ -term reduces to the usual anisotropic spin exchange  $\sim [\tau_i^y \tau_j^y - \tau_i^x \tau_j^x - \tau_i^z \tau_j^z]$  in the two-orbital limit. Analogously,  $\mathcal{H}_{\hat{y}}$  reduces to the two-component  $(p_y, p_z)$ -orbital model with  $(\lambda^6, \lambda^7, \lambda^y)$  acting as another set of Pauli matrices  $\boldsymbol{\mu}$  with the DM interaction,

$2t\gamma \sum_i \hat{y} \cdot [\boldsymbol{\mu}_i \times \boldsymbol{\mu}_{i+\hat{y}}]$ . Such Pauli matrix description (effective spin-1/2 model) must give way to the full Gell-Mann matrix formalism shown in Eq. (2) once  $\mathcal{H}_{\hat{x}}$  and  $\mathcal{H}_{\hat{y}}$  are combined in the two-dimensional lattice. A similar Gell-Mann matrix expression appeared in the low-energy theory of hard-core three-component bosons confined in the optical lattice [8] where, however, the influence of ISB on the superexchange process was not examined.

We can further organize the Gell-Mann matrix-based Hamiltonian of Eq. (2) with the following observation. First we define, formally, the  $S = 1$  spin operator  $\mathbf{S} = (S^x, S^y, S^z) = (\lambda^7, -\lambda^5, \lambda^2)$  satisfying  $(S^\alpha)_{\beta\gamma} = -i\varepsilon_{\alpha\beta\gamma}$ . Then the remaining five Gell-Mann matrices are generated as

$$\begin{aligned}
Q^{xy} &= S^x S^y + S^y S^x = -\lambda^1, \\
Q^{yz} &= S^y S^z + S^z S^y = -\lambda^6, \\
Q^{zx} &= S^z S^x + S^x S^z = -\lambda^4, \\
Q^{x^2-y^2} &= [S^x]^2 - [S^y]^2 = -\lambda^3, \\
Q^{3z^2-r^2} &= \frac{1}{\sqrt{3}}(2[S^z]^2 - [S^x]^2 - [S^y]^2) = \lambda^8, \quad (4)
\end{aligned}$$

which are precisely the five nematic operators often employed in the discussion of quantum nematic order in

spin-1 models [9, 10]. Our Hamiltonian (2) thus embodies complex exchange interactions among (pseudo-)spin and nematic order parameters.

We perform the mean-field analysis of the phase diagram based on Eq. (2), by making replacement of the operator  $\lambda_i^\alpha$  with its average,  $\langle \lambda_i^\alpha \rangle$ , in the Hamiltonian. Assuming site-factorized wave function

$$|\psi_i\rangle = A_i^x |X_i\rangle + A_i^y |Y_i\rangle + A_i^z |Z_i\rangle \quad (5)$$

with the three complex coefficients satisfying  $|A_i^x|^2 + |A_i^y|^2 + |A_i^z|^2 = 1$ , the many-body wave function becomes the direct product  $|\psi\rangle = \prod_i \otimes |\psi_i\rangle$ . In this scheme one can choose  $A_i^x$  real without loss of generality and parameterize the coefficients generally as  $A_i^x = \cos \alpha_i$ ,  $A_i^y = e^{i\gamma_i} \sin \alpha_i \cos \beta_i$ ,  $A_i^z = e^{i\delta_i} \sin \alpha_i \sin \beta_i$ . The mean-field Hamiltonian being a function of the four angles  $(\alpha_i, \beta_i, \gamma_i, \delta_i)$  per site can be minimized by the Monte Carlo (MC) annealing method. We enrich the phase diagram of the model (2) by introducing one more parameter  $A$  to replace  $t\gamma \rightarrow tA\gamma$ . Varying the strength of  $A$  independently of  $\gamma$  permits more control of the strength of orbital DM exchange over the other interactions in the model. The zero-temperature phase diagram spanning  $(\gamma, A)$  is shown in Fig. 1. Everywhere we find  $\langle \mathbf{S}_i \rangle = 0$ . Recall  $(S^\alpha)_{\beta\gamma} = -i\varepsilon_{\alpha\beta\gamma}$  is pure imaginary, while our numerical MC search only turned up real-valued wave functions,  $\gamma_i = \delta_i = 0$ . The remaining six Gell-Mann matrices, including the unit matrix, can be organized into two groups  $\mathbf{T}_i^x = (\lambda_i^4, \lambda_i^5, \lambda_i^x)$  and  $\mathbf{T}_i^y = (\lambda_i^6, \lambda_i^7, \lambda_i^y)$  as was previously considered for  $\mathcal{H}_{\hat{x}}$  and  $\mathcal{H}_{\hat{y}}$ , respectively. The ground state mean-field configuration was Fourier analyzed  $\mathbf{T}_\mathbf{k}^n = \sum_i \langle \mathbf{T}_i^n \rangle e^{-i\mathbf{k} \cdot \mathbf{r}_i}$  ( $n = x, y$ ) in search of modulated structures with long periods that often appears with the DM interaction.

When  $\gamma = 0$  the Hamiltonian  $H_{\hat{x}} + H_{\hat{y}}$  is expressed in terms of two commuting matrices,  $(\lambda_i^x, \lambda_i^y)$ , with the ground state given by alternate occupations of  $p_x$  and  $p_y$  orbitals on the square lattice. This phase, called the antiferro-orbital (AFO) state, dominates the small- $\gamma$  region of the phase diagram. For  $A$  small and  $\gamma$  increasing beyond a critical value, one finds a first-order transition into a ferro-orbital (FO) state with  $p_z$ -orbital occupation at every site. Inhabiting the large- $A$ , intermediate- $\gamma$  region is the  $\langle 11 \rangle$  helical (H) phase that we found to be well described by

$$\begin{aligned} \langle \mathbf{T}_i^x \rangle &= (\sin 2[\mathbf{k} \cdot \mathbf{r}_i + \alpha_0] \sin[\mathbf{k} \cdot \mathbf{r}_i + \beta_0], 0, \\ &\quad \cos^2[\mathbf{k} \cdot \mathbf{r}_i + \alpha_0] - \sin^2[\mathbf{k} \cdot \mathbf{r}_i + \alpha_0] \sin^2[\mathbf{k} \cdot \mathbf{r}_i + \beta_0]), \\ \langle \mathbf{T}_i^y \rangle &= (\sin^2[\mathbf{k} \cdot \mathbf{r}_i + \alpha_0] \sin 2[\mathbf{k} \cdot \mathbf{r}_i + \beta_0], 0, \\ &\quad \sin^2[\mathbf{k} \cdot \mathbf{r}_i + \alpha_0] \cos 2[\mathbf{k} \cdot \mathbf{r}_i + \beta_0]), \end{aligned} \quad (6)$$

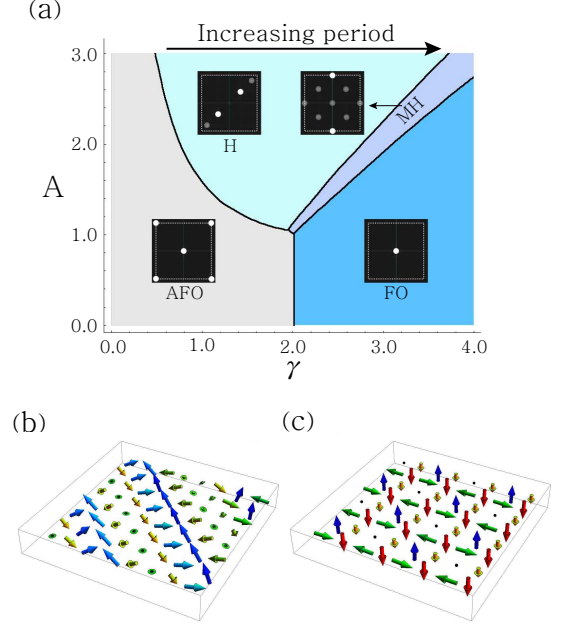


FIG. 1: (color online) (a) Zero-temperature phase diagram of the orbital model in Eq. (2) with  $t = 1$ . Phase boundaries are obtained on the basis of extensive MC simulation and variational energy calculation. Orbital configurations are abbreviated as AFO (antiferro-orbital), FO (ferro-orbital), H (helical), and MH (multiple helical). Bragg patterns for each phase are schematically shown with dotted line denoting the first Brillouin zone boundary. In H, the period increases continuously as  $\gamma$  becomes larger. Bragg spots at  $(k, k)$  and  $(3k, 3k)$  are both present in H. Real space spin configurations in terms of  $\langle \mathbf{T}_i^x \rangle$  for (b) H and (c) MH phases are depicted.

$\mathbf{k} = (k, k)$ . Variational calculation of the minimum energy with respect to  $k$  and the relative phase angle  $\alpha_0 - \beta_0$  confirms that the period  $2\pi/k$  is increased with  $\gamma$  in  $A > 1$  region from 2 to  $> 6$  before it is supplanted by another intricate phase taking place between H and FO. The new phase, indicated as MH (multiple helical) in Fig. 1) is constructed as the equal-weight superposition of two helices with  $\mathbf{k}_1 = \pm(\pi/2, \pi/2)$  and  $\mathbf{k}_2 = \pm(\pi/2, -\pi/2)$ , as well as two asymmetric peaks at  $(\pi, 0)$  and  $(0, \pi)$ .

The problem of including the physical spin in the three-orbital model proved too complicated, but there is an alternative to study much of the same physics, hopefully, in a more tractable two-orbital model. Write down the two orbitals carrying spin  $\sigma$  as  $a_{i,\sigma}$  and  $b_{i,\sigma}$ , and the spinor  $\psi_{i,\sigma} = \begin{pmatrix} a_{i,\sigma} \\ b_{i,\sigma} \end{pmatrix}$ . The two-orbital spin model features hopping among the same orbitals with amplitudes  $t_a$  and  $t_b$ , respectively, and the inter-orbital hopping proportional to the ISB parameter  $\gamma$ :  $H_t = \sum_{\langle ij \rangle, \sigma} \psi_{i,\sigma}^\dagger (t_1 + i\gamma\tau_y + t_2\tau_z) \psi_{j,\sigma}$ . The Pauli matrix  $\boldsymbol{\tau}$  is introduced to span the  $2 \times 2$  matrix and  $t_1 = (t_a + t_b)/2$ ,  $t_2 = (t_a - t_b)/2$ . Time-reversal symmetry is preserved for this model. Implementing separate unitary rotations

for  $\psi_i$  and  $\psi_j$ ,  $\psi_i \rightarrow e^{i\theta\tau_y/2}\tilde{\psi}_i$ ,  $\psi_j \rightarrow e^{-i\theta\tau_y/2}\tilde{\psi}_j$  [11],  $H_t$  becomes

$$\sum_{\langle ij \rangle, \sigma} \left( t\tilde{\psi}_{i,\sigma}^\dagger \tilde{\psi}_{j,\sigma} + t_2\tilde{\psi}_{i,\sigma}^\dagger \tau_z \tilde{\psi}_{j,\sigma} + h.c. \right) \quad (7)$$

for each adjacent pair  $\langle ij \rangle$ , and  $t(\cos\theta, \sin\theta) = (t_1, \gamma)$ . Superexchange calculation is most conveniently carried out in the new basis, assuming the quarter-filling (one

electron per site), with the resulting spin-orbital exchange Hamiltonian for each pair  $\langle ij \rangle$ :

$$\mathcal{H}_{ij} = P_{ij}\boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j + Q_{ij}\sigma_i^z \sigma_j^z + R_{ij}. \quad (8)$$

Here  $\boldsymbol{\sigma}_i$  is the spin operator at site  $i$ . The orbital operators  $\boldsymbol{\tau}_i$  appear in the complex form,

$$\begin{aligned} P_{ij} &= \frac{C}{2} \left( (t^2 + t_2^2)(\tilde{\tau}_i^z \tilde{\tau}_j^z + 1) + 2tt_2(\tilde{\tau}_i^z + \tilde{\tau}_j^z) \right) + \frac{B}{4}(t^2 - t_2^2)(\tilde{\tau}_i^+ \tilde{\tau}_j^- + \tilde{\tau}_i^- \tilde{\tau}_j^+), \\ Q_{ij} &= \frac{(A-B)}{2} \left( (t^2 + t_2^2)(\tilde{\tau}_i^z \tilde{\tau}_j^z - 1) + \frac{(t^2 - t_2^2)}{2}(\tilde{\tau}_i^+ \tilde{\tau}_j^- + \tilde{\tau}_i^- \tilde{\tau}_j^+) \right), \\ R_{ij} &= \frac{(t^2 + t_2^2)}{2}(A + B - C)\tilde{\tau}_i^z \tilde{\tau}_j^z - Ctt_2(\tilde{\tau}_i^z + \tilde{\tau}_j^z) + \frac{A}{4}(t^2 - t_2^2)(\tilde{\tau}_i^+ \tilde{\tau}_j^- + \tilde{\tau}_i^- \tilde{\tau}_j^+), \end{aligned} \quad (9)$$

with  $A = t^2/(U-3J_H)$ ,  $B = t^2/(U-2J_H)$ , and  $C = t^2/U$ . The Hund's exchange  $J_H$  is included in the present calculation. All the orbital operators  $\tilde{\boldsymbol{\tau}}_i$  and  $\tilde{\boldsymbol{\tau}}_j$  are written in the transformed basis  $\tilde{\psi}_i$  and  $\tilde{\psi}_j$ , and to recover the form in the original orbital basis one needs the rotation  $\tilde{\tau}_i^z = \tau_i^z \cos\theta - \tau_i^x \sin\theta$ ,  $\tilde{\tau}_j^z = \tau_j^z \cos\theta + \tau_j^x \sin\theta$ , and  $\tilde{\tau}_i^x = \tau_i^x \cos\theta + \tau_i^z \sin\theta$ ,  $\tilde{\tau}_j^x = \tau_j^x \cos\theta - \tau_j^z \sin\theta$ . As a result  $\tilde{\boldsymbol{\tau}}_i \cdot \tilde{\boldsymbol{\tau}}_j$  and  $\tilde{\tau}_i^z \tilde{\tau}_j^z$  in Eq. (9) become

$$\begin{aligned} \cos 2\theta \boldsymbol{\tau}_i \cdot \boldsymbol{\tau}_j + 2\sin^2\theta \tau_i^y \tau_j^y + \sin 2\theta (\hat{y} \cdot \boldsymbol{\tau}_i \times \boldsymbol{\tau}_j), \\ \cos^2\theta \tau_i^z \tau_j^z - \sin^2\theta \tau_i^x \tau_j^x + \frac{1}{2} \sin 2\theta (\hat{y} \cdot \boldsymbol{\tau}_i \times \boldsymbol{\tau}_j), \end{aligned} \quad (10)$$

respectively. There is again the orbital version of the DM interaction,  $\hat{y} \cdot \boldsymbol{\tau}_i \times \boldsymbol{\tau}_j$ , with strength proportional to  $\sin 2\theta \sim \gamma$ .

A simpler form follows from taking  $J_H = 0$ ,

$$\begin{aligned} \mathcal{H}_{ij} &= (\boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j + 1)(\tilde{\boldsymbol{\tau}}_i \cdot \tilde{\boldsymbol{\tau}}_j + 1) + \frac{2t_2}{t}(\boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j - 1)(\tilde{\tau}_i^z + \tilde{\tau}_j^z) \\ &\quad + \left(\frac{t_2}{t}\right)^2 (\boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j + 1)(2\tilde{\tau}_i^z \tilde{\tau}_j^z - \tilde{\boldsymbol{\tau}}_i \cdot \tilde{\boldsymbol{\tau}}_j + 1), \end{aligned} \quad (11)$$

where the overall exchange energy  $J = t^2/2U$  is taken to unity. The first term possesses the well-known SU(4) symmetry [12], broken down to SU(2)×U(1) by terms of order  $t_2/t$  and  $(t_2/t)^2$  in the above. Microscopically nonzero  $t_2$  arises from different hopping amplitudes of  $\sigma$ -bonding and  $\pi$ -bonding orbitals on the lattice. One can see the qualitative effects of the symmetry-breaking terms within the mean-field picture. The interaction linear in  $t_2/t$  favors a pair of adjacent spins to be antiparallel,  $\langle \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j \rangle - 1 < 0$ , therefore the orbital opera-

tor  $\tilde{\tau}_i^z$  acquires an overall Zeeman field that favors ferro-orbital order:  $\langle \tilde{\tau}_i^z \tilde{\tau}_j^z \rangle > 0$ . With the additional conditions  $\langle \tilde{\tau}_i^x \tilde{\tau}_j^x \rangle = \langle \tilde{\tau}_i^y \tilde{\tau}_j^y \rangle = 0$  one can prove the orbital correlations in the original basis:  $\langle \hat{y} \cdot \boldsymbol{\tau}_i \times \boldsymbol{\tau}_j \rangle = \langle \tilde{\tau}_i^z \tilde{\tau}_j^z \rangle \sin 2\theta$ . In the case when the  $(t_2/t)^2$  term dominates one expects ferro-spin order  $\langle \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j \rangle + 1 > 0$  with easy-plane anisotropy for the orbital  $\boldsymbol{\tau}_i$ :  $\langle \tilde{\tau}_i^z \tilde{\tau}_j^z \rangle = 0$ ,  $\langle \tilde{\tau}_i^+ \tilde{\tau}_j^- \rangle \neq 0$ , implying  $\langle \hat{y} \cdot \boldsymbol{\tau}_i \times \boldsymbol{\tau}_j \rangle = (1/2)\langle \tilde{\tau}_i^+ \tilde{\tau}_j^- \rangle \sin 2\theta$ . The ground state of Eq. (11) is thus expected to show strong chiral orbital order  $\langle \hat{y} \cdot \boldsymbol{\tau}_i \times \boldsymbol{\tau}_j \rangle \neq 0$  throughout the wide parameter range of  $t_2/t$ . A full quantum-mechanical evaluation of the phase diagram will be carried out in a future publication.

Unlike the spin-DM interaction in materials, the governing factor  $\gamma$  responsible for the orbital analogue of DM exchange can be imposed externally by the electric field in a controlled manner. Interesting quantum-orbital phases and transitions between them may be observed in a thin-film multi-orbital system subject to perpendicular electric field.

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